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A REVIEW OF APPLICATIONS OF MASS SPECTROMETRY TO LOW LEVEL RADIONUCLIDE METROLOGY

bу

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A REVIEW OF APPLICATIONS OF MASS SPECTROMETRY TO LOW LEVEL RADIONUCLIDE METROLOGY*

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ABSTRACT

Radiation counting of nuclear decay events is the most commonly used method of measuring radionuclide concentrations. However, some radionuclides, such as transuranics: 235U, 236U, 240Pu, 241Pu, 242Pu, and the fission products: 99Tc and 135Cs can be analyzed by mass spectrometry with greater sensitivity, ease, and/or accuracy. Mass spectrometry also offers an independent means of measuring radionuclide concentrations which can be valuable as a comparison with radiation counting.

Current capabilities of mass spectrometry for analysis of selected environmental radionuclides as well as projected developments for the near future are summarized. Comparisons of cost, sensitivity, and accuracy (with a particular emphasis on sensitivity) are made for those radionuclides of most interest to environmental studies and for those most likely to lend themselves to mass spectrometric analysis. In some cases where there are presently no mass spectrometric methods which are competitive with radiation counting, limiting factors are defined, and the possibility of future developments is discussed.

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INTRODUCTION

There are numerous reasons for measuring low levels of radionuclides. Included among them are tracer experiments of several types (biological, medical, oceanographic, and meteorological to name a few), radioactive decay methods for archeological and geological age determination, monitoring of nuclear effluents, and studies of transport modes of radioisotopes to help determine and predict long-term environmental impact of nuclear facilities.

Regardless of the application, it is desirable that the technique for analysis of a radionuclide be sensitive, accurate, and give real-time results at modest cost. Untortunately no single analysis technique can obtain all of these goals for all applications. Each technique has its own strengths and weaknesses. This work is a comparison of mass spectrometric analysis techniques which are currently in use or easily adaptable for measurement of low levels of radionuclides with the more traditional radioactive decay counting techniques.

A general comparison of mass spectrometry and counting techniques is presented in the theory section of this work. To illustrate the general comparisons made in the theory section, techniques for analysis of tritium, carbon-14, cesium, technetium-99, iodine-129, and plutonium are discussed in greater detail. These radionuclides were chosen for further discussion because they are of significant interest and because they demonstrate specifically the relative merits of mass spectrometry and counting for a variety

of types of nuclides. Table 1 follows the examples and is a more inclusive, although by no means all inclusive, comparison of mass spectrometric and radioactive decay counting sensitivities for a number of radionuclides.

The final section of this work is a brief discussion of possible future developments in mass spectrometry which should be applicable to radionuclide metrology.

THEORY

The sensitivity of a technique is one of the most important factors used to evaluate the relative merit of that technique. This is especially true for analysis methods for low-level radionuclide metrology. For this reason, and to limit the scope of this discussion, sensitivity will be the primary factor used to compare counting and mass spectrometric techniques in this review. There are many different ways to quantify sensitivty. Terms such as limit of detection, limit of determination, minimum detectable amount, and ultimate detection sensitivity abound in the literature. Also, the authors may or may not define what they mean by these terms. For these reasons it is often difficult to make comparisons of the sensitivity of different techniques. For the sake of brevity, simplicity and uniformity the term "limit of detection" will be used throughout the text with the following definition. The limit of detection of an instrument (be it a mass spectrometer or a nuclear decay counting device) is the smallest quantity of sample (the units of Bq are used throughout the text)

required to produce a signal S sufficiently greater than the background B of a blank sample to result in an assigned measurement error ε equal to one half of the net sample signal (S-B). That is $\varepsilon = 1/2$ (S-B). More involved discussions of sensitivity occur in the literature (see for instance References 1-5. However, for the comparisons of very different techniques made in this work this simple definition is adequate. An attempt has been made to use the same definition and units for limit of detection throughout this review. Thus, assumptions and conversions often had to be made to calculate the limit of detection for a particular technique.

In order to make some general statements about the relative sensitivity of mass spectrometry and radioactive decay counting, the limit of detection for the two techniques is calculated below using the above definition and assuming normal counting statistics. The limit of detection (LD) for a radioactive decay counter can be calculated to be:

$$LD = \frac{2}{\varepsilon_c \beta t_c} \left(1 + \sqrt{1 + 2B \epsilon_c + \sigma_B^2 \epsilon_c^2} \right), \quad (Equation 1)$$

where LD is expressed in Bq, ϵ_c is the counter efficiency expressed as a fraction, β is the branching ratio for the measured radiation expressed as a fraction, t_c is the counting time in seconds, B is the background count rate for the detector; and σ_B is the error in the measurement of the background.

Similarly the limit of detection for a mass spectrometer $(LD_{\mathfrak{m}})$ can be written as:

$$LD_{m} = \frac{2 \ln (0.5)}{\varepsilon t_{1/2}} \left(1 + \sqrt{1 + 2B_{m}t_{m} + \sigma_{B}^{2}t_{m}^{2}} \right), \quad (Equation 2)$$

where the limit of detection is given in Bq, $t_{1/2}$ is the half-life of the radionuclide in seconds, t_m is the mass spectrometer run time, B_m is the mass spectrometer background count rate, σ_B is the measurement error of the background, and ϵ is the overall efficiency of the mass spectrometer expressed as a fraction. The mass spectrometer efficiency (ϵ) is the product of the ionization efficiency, the transmission efficiency and the detector counting efficiency. For most modern mass spectrometer systems with pulse counting capabilities, the transmission efficiency and detector efficiency are virtually unity.

The mass spectrometer efficiency (E) is most strongly influenced by the ionization efficiency which can vary widely depending on the ion source and the element which is being analyzed. The thermal ionization source, one of the most commonly used ion sources is an excellent example of the dependence of the ionization efficiency on sample composition. This source is based on the fact that when neutral atoms with an ionization potential I are heated on a hot metallic filament surface of work function W and temperature T, there is a probability that ions will evaporate in addition to neutral atoms. The ratio of the number of positive ions to the number of neutral atoms evaporated is given by:

$$\frac{n_{+}}{n_{0}} = \frac{1 - r_{+}}{1 - r_{0}} \frac{g_{+}}{g_{-}} \exp \left[e(W-I)/kT\right], \qquad (Equation 3)$$

where r_+ , r_0 , g_+ and g_0 are constants specific the filament material, and the thickness and composition of the solid sample.⁶ Except for noting that the terms r_+ , r_0 , g_+ , and g_0 are generally not well defined, further discussion of these terms is beyond the scope of this work. Further discussion may be found in Reference 6 and references cited therein. In spite of the variability in the constant of proportionality the formula

$$\frac{n_{+}}{n_{0}} \propto \exp \left[e(W-I)/kT\right]$$
 (Equation 4)

can be used as a rough guide when comparing the relative ionization efficiencies of various elements on particular filament materials and at varying temperatures. Thus, it can be seen that as a general rule those elements with lower ionization potentials are more efficiently ionized. An example of this is discussed later when comparing the relative mass spectrometric sensitivity for technetium and plutonium.

For those elements with a high electron affinity, the thermal source can be converted to an electron affinity or negative surface ionization source simply by reversing the source polarity. The ratio of negative ions to neutral atoms, n_/n_0, can be obtained by replacing W-I in Equation 4 with A-W where A is the electron affinity of the sample element. The use of this type source for the analysis of iodine is discussed later in the text.

when comparing sensitivities, another important difference between Equations 1 and 2 is the half-life term which appears in the denominator of Equation 2. This term is a result of the fact that the mass spectrometer measures the parent isotope and not the decay event. Because of this half-life term, mass spectrometry should become a preferred technique for radioisotopes with long nalf-lives. It is possible to make some simple assumptions and calculate the minimum half-life for which the mass spectrometer can be a competitive technique. To illustrate this, assume a hypothetical alpha counting technique with the following capabilities:

 ϵ_c = 0.25, t_c = 6 x 10⁵ sec (~1 week), B = 4 x 10⁻⁶ cps, and β = 1 and a hypothetical mass spectrometer with the following capabilities:

 ϵ = 0.005, t_m = 1000 sec, B_m = 0.002 cps.

In both cases assume that σ_B is negligibly small. By equating the sensitivity of the two techniques given by Equations 1 and 2, substituting the values listed above and solving for $t_{1/2}$, the half-life for which the sensitivity of mass spectrometry and decay counting is approximately equal is 230 days. These two hypothetical techniques are approximately the current state of the art capabilities for the plutonium isotopes.

Another term in Equation 2 which merits further discussion is the background term. In the above example, a typical mass spectrometer counting system background was assumed. Other factors may result in an increase in the background B_m and/or the background measurement error σ_B , which in turn will result in an increase in the limit of detection.

The two primary factors which may increase $B_{\rm m}$ are the existence of isobars in the sample which cannot be resolved and the tailing of large neighboring mass peaks into the mass region of the isotopes of interest. Interferences due to isobars are usually limited by chemically separating the sample and by judiciously choosing construction materials for the mass spectrometer components. The tailing problem can be limited to a certain extent by constructing mass spectrometers with a high abundance sensitivity, typically by placing selection steps in series, which results in a multiplication of abundance sensitivities for each step.

There are several practical considerations which must be weighed when considering the use of the mass spectrometer for measuring radionuclides. Among them are: the possibility of contaminating an expensive instrument, the cost of the analysis (the element of interest must usually be chemically separated from its sample matrix and the initial instrument expense is nontrivial), n isotope for yield tracing and the availability of a mass spectrometer suitable for analyzing the particular element of interest.

There are cases where yield tracing isotopes are not used. It is often sufficient information to measure the ratios of isotopes in a sample. This may be the case when one of the isotopes is naturally occurring either at a known concentration or measurable concentration. The measurement of iodine-129 by mass spectrometry or high energy mass spectrometry (accelerator techniques) is an

example of a situation where the stable concentration is determined by standard analytical techniques and the radioactive species concentration is derived from the ratio of radioactive species to the stable isotopes. Another example where the isotope ratio is sufficient information is the case where normal counting techniques cannot resolve two isotopes, but can be used to measure the total concentration of the two isotopes. In this case the mass spectrometer can be used to determine the relative contribution of each isotope to the overall concentration. Uranium 233, 234, uranium 235, 236, and plutonium 239, 240 are examples of pairs of isotopes which are not normally resolved by alpha spectrometry, but can easily be resolved by mass spectrometry.

DISCUSSION

Plutonium

Mass spectrometric analysis of plutonium is a good example of the capability of mass spectrometry for the detection of low levels of a long-lived radionuclide. Lagergren has reported a limit of detection of 4 x 10⁻¹⁸ g for plutonium.⁷ Thus the limit of detection for plutonium by Lagergren's technique can be calculated to vary from 80 μBq for ²³⁶Pu to 2.7 x 10⁻⁶ μBq for ²⁴⁴Pu. This compares very favorably with the alpha counting limit of detection quoted by Peuser et al. of 370 μBq.⁸ Also, from a practical standpoint, plutonium lends itself to mass spectrometric analysis. There are very few isobar interferences, ²³⁸U being a notable exception. There is a large choice of isotopes which can be used

for yield tracing. Since most of the isotopes are so long-lived and the instrument is sensitive to such low levels, small enough samples can be used so that there is little concern of contamination.

Technetium

Technetium has two long-lived man-made isotopes, 97 Tc and 99 Tc with half-lives of 2.6 x 106 years and 2.1 x 105 years, respectively. Technetium-99 is a fission product of some environmental concern. For this reason, several methods to analyze for 99 Tc have been developed. Because 99 Tc is a pure beta emitter with a long half-life, it is not easily measured using conventional counting techniques. On the other hand, there are several reasons that 99 Tc lends itself to mass spectrometric analysis. There are no stable isotopes of technetium which can limit sensitivity because of mass spectrometer abundance sensitivity limitations. 97 Tc can be used as a yield determinant. There is only one stable isobar, 99 Ru, which can be chemically separated. Anderson has reported a method of chemical separation and mass spectrometric analysis for 99Tc which is capable of a limit of detection of 600 µBq. 9 The major difference between technetium and plutonium which causes the mass spectrometer's sensitivity to be less for technotium is that the ionization potential of technetium is much higher than plutonium's ionization potential. As mentioned earlier, this causes a much lower ionization efficiency when using standard surface ionization ion sources.

Cesium

Cesium has three isotopes which are radioactive and for which there is the possibility of measurement using mass spectrometry, $134_{\rm Cs}$, $135_{\rm Cs}$, and $137_{\rm Cs}$. The half-lives of these isotopes are: 134 Cs - 2.06 years, 135 Cs - 3 x 106 years, and 137 Cs - 30.2 years. Decay chains for both 134Cs and 137Cs include gamma ray radiations. Due to the relative ease of measuring and identifying these gamma rays, mass spectrometry is seldom considered as a method for analysis of these isotopes in natural matrices. Also, due to the short half-lives, there is little advantage in measuring the parent atom using the mass spectrometer. The long-lived isotope, 135Cs, is a pure beta emitter. Since beta radiation is more difficult to measure using radioactive decay methods, and because of the very long half-life, 135 Cs is a very likely candidate for analysis by mass spectrometry. Another fact that makes mass spectrometry attractive for analysis of 135 Cs is that cesium is very efficiently ionized using conventional surface ionization sources (cesium has a very low ionization potential). There is one stable isotope of cesium (mass = 133) which may occur in sufficient quantity in a sample to limit sensitivity due to the lack of abundance sensitivity of the mass spectrometer. Also there is the isobar 135 Ba which must be chemically removed from the cesium sample to permit sensitive analysis. Brauer and Ballou have reported measurement of 135Cs in air samples near a nuclear facility. 10 Estimates can be made to calculate approximate mass spectrometric sensitivity for

135 Cs. Using results for mass spectrometric analysis of other cesium isotopes 11,12 and assuming a concentration of the stable isotope 133 in the sample to be approximately equal to the concentration in the earth's crust, we can calculate that it is possible to measure concentrations as small as 0.2 µBq of 135 Cs/g of soil or similar sample matrix. The major limiting factor is the abundant stable cesium isotope and the limit of abundance sensitivity which can be obtained with a conventional mass spectrometer.

Carbon-14

Sensitive analysis for 14C by conventional mass spectrometry is difficult because of the highly abundant stable isotopes in most samples of interest. Also carbon is not easily ionized, and it forms molecular species during acceleration in the ion source. These molecular species can form ions with masses nearly equal to the species containing the 14C isotope. There are also significant isobar interferences, in particular the very abundant 14N isotope. In spite of all of these adversities, Hedges 13 proposes that detection of 14C at modern abundances may be possible. Since much of the interest in measurement of 14C is for dating purposes, more sensitive techniques are often required. One such technique is known as high energy mass spectrometry. This technique utilizes nuclear accelerators (cyclotrons and tandem electrostatic accelerators) as mass spectrometers. Many of the difficulties of conventional mass spectrometry are overcome by utilizing the high energies and negative ions available in accelerator based systems.

Using a nuclear accelerator, Gove claims 10⁵ times more sensitivity than conventional counting techniques. 14 Unfortunately, running an accelerator is significantly more costly than operating a Geiger-Müller tube. For this reason, the high energy mass spectrometer is usually reserved for those samples which cannot be analyzed by conventional counting techniques, primarily due to sample size limitations.

Tritium

Tritium is an isotope which is extremely difficult to measure at low levels by mass spectrometry. There are isobar interferencess, molecular forms which interfere, and highly abundant stable isotope, all of which present problems for conventional mass spectrometric analysis for tritium. Decay counting methods for tritium are well developed and are sensitive enough for most applications. Quoted limits of detection vary from 0.2 Bq for liquid scintillation to 0.0004 Bq for proportional counting to 40 µBq for proportional counting with electrolytic enrichment. 15,16

A competitive mass spectrometric technique has been developed by Clarke, Jenkins, and Top. 17 The technique utilizes a very sensitive static noble gas mass spectrometer for analysis of accumulated 3He, the decay product of tritium. Clark, Jenkins, and Top have quoted a limit of detection of 370 µBq using this technique.

Iodine

Iodine has several radioactive isotopes. However, only ¹²⁹I has a half-life long enough to lend itself to mass spectrometric analysis. Iodine has a very high ionization potential and

therefore does not form significant quantities of positive ions in a conventional positive surface ionization source. However, recent work by J. E. Delmore has shown promising results for analysis of $^{129}{}_{\rm I}$ using negative surface ionization. $^{18}{}$ He has reported the capability of measuring $^{127}{}_{\rm I}$ to $^{129}{}_{\rm I}$ ratios as large as 10^8 . This capability is sufficient to measure modern concentrations of $^{129}{}_{\rm I}$ as reported by Brauer. $^{19}{}_{\rm I}$ The results of Delmore were achieved using large samples of iodine (0.85 $\mu{}_{\rm B}$). The limit of detection for $^{129}{}_{\rm I}$ using this technique with a large iodine sample can be calculated to be at least as small as 0.05 Bq, which compares favorably with gamma ray spectrometry (0.02 Bq), but not with neutron activation (0.04 $\mu{}_{\rm B}{}_{\rm Q}$) reported by Brauer. $^{15}{}_{\rm I}$ Judging by the high ionization efficiency Delmore quoted in his work, much smaller samples of iodine could be analyzed, which would in turn reduce the limit of detection.

Another method for analysis of iodine is described by Boulos, Ballard, and coworkers. 20,21 They utilized a mass spectrometer in conjunction with neutron activation. The mass spectrometer is used for the measurement of accumulated xenon isotopes after neutron activation of the iodine isotopes:

$$127_{I(n,2n)}$$
 126_{I} $\frac{\beta^{-}}{13d}$ 126_{Xe}

$$^{129}I(n,\gamma)^{130}I \frac{\beta^{-}}{124 \text{ hr}} \quad ^{130}Xe$$

Using this technique, they have reported limits of detection as low as 120 µBq. The limitation of sensitivity by this technique and Brauer's technique is the triple neutron capture of ¹²⁷I. The discrepancy in sensitivity between Brauer and Ballard is a function of the initial sample sizes required by the techniques.

NEW TECHNOLOGIES

Two significant advances have occurred in recent years which have possible applications to radionuclide metrology. One has already been eluded to during the discussion of ¹⁴C, that is high energy mass spectrometry. This technique has already proven useful for analysis of ¹⁰Be, ²⁶Al, ³²Si, ³⁶Cl, and ¹²⁹I. See for instance Elmore et al., ²² Müeller et al., ²³ Rucklidge et al., ²⁴, Gove, ¹⁴ and Hedges ¹³.

The other advance is in the area of resonance ionization spectroscopy and lasers, first developed by Hurst and coworkers.²⁵
This technique utilizes the very narrow linewidths and high powers available with today's lasers to selectively excite and subsequently ionize single atoms. Hurst and coworkers have proposed five schemes of ionization shown in Figure 1. With these schemes they can ionize any element except helium and neon using presently available laser systems. For certain applications, in particular the lighter elements where isotope shifts in the atomic states are large, this method is isotopically selective and may have direct application to measurement of radionuclides. Those elements for which there is little isotopic selectivity may lend themselves to

analysis by a combination of resonance ionization spectroscopy and mass spectrometry. This combination may be used to improve ionization efficiency of elements not previously efficiently ionized, it may be used as a substitute or enhancement of chemical separation, or the isotope selectivity of resonance ionization spectroscopy may be used to enhance the abundance sensitivity of mass spectrometry.

CONCLUSION

Mass spectrometry has applications to low-level radionuclide metrology. In particular, it can be the method of choice for very low-level measurements of very long-lived radionuclides. Because of isobar and chemical interferences, it is usually necessary to chemically separate the element of interest. For this reason and due to the initial expense of the mass spectrometer, mass spectrometry is relatively more expensive than some decay counting techniques. However, the mass spectrometer can be cost competitive to other techniques which require chemical separation. It is for this reason, and often because of the increased sensitivity afforded by mass spectrometry, that mass spectrometry is sometimes chosen rather than beta and alpha counting techniques. Even if mass spectrometry is more costly or less sensitive, there are some situations which may require an independent and/or accurate measurement for calibration or confirmation; here again, mass spectrometry may be the technique of choice.

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TABLE 1: SENSITIVITY COMPARISON, DECAY COUNTING VS. MASS SPECTROMETRY

7	A	ELEMENT	HALF LIFE (YEARS)	MAJOR Radiations	COUNT. SENS. (BQ)	COUNTING METHOD	REF.	M. S. SENS. (BQ)	M. S. METHOD	LIMITA- TIONS	REF.
1	3	HYDROGEN	1.23E+01	6-	1.9E=01	LS	15	3.7E-04	SG	C, I, S	17
1	3	HYDROGEN	1.236-01	8-	3.7E-03	PC ,	15,14				
1	3	HYDROGEN	1.23E+01	8=	3.7E-05	EE+PC	15,16		:		
6	14	CARBON	5.73E+03	8-	1.7E-05	PC(MINI)	26	3.06-09	HE	C, I, S, IE	13,14
56	55	IRON	2.60E+00	6 .	4.4E-02	PC	27			T,5,1,0	
36	85	KRYPTON	1.08E+01	B≃,G	6.0E-04	PC	31	8.0E+01	5 G	5, C	EST. **
43	99	TECHNETIUM	2.00E+05	8-	7.4E-03	PC	15	6.0E-04	SI		9
53	129	IODINE	1.70E+07	8-,E-,G	1.8E-02	GS	15	1.8E-13	HE	8	24 #
53	129	IODINE	1.70E+07	B-,E-,G	3.7E-08	NAA	15	5.0E-02	NSI	8,c	18
- 55	134	CESIUM	2.06E+00	8-,G	6.0E-03	GS	33			T,8,1,\$	
55	135	CESIUM	3.00E+06	8-		•		2.0E-01	SI	3	EST. ***
55	137.	CESIUM	3.00E+01	B-,EC,G	7.0E-03	GS .	28			S,8,T,I	
63	152	EUROPIUM	1.275+01	8-,6-,8+,9		0.5				3,3,1,7	1
63	154	EUROPIUM	1.602+01	B-,E-,G		G5		,•		5,4,1,T	
63	155	EUROPIUM	1.80E+00	B-, E-, G	1.0E-02	GS	26			T, S, S, I	
82	210	LEAD	2.04E+01	B-,G	1.6E-01	GS	58	7.0E-01	51	3, IE, T, \$	EST.***
82	210	LEAD	2.04E+01	B-,G	1.56-03	PC	30				
92	233	URANIUM	7.20E+01	A,G,E-	5.3E-01	A5 *	32	1.5E-01	51		29
92	234	URANIUM	2.475+05	A,G	7.6E-04	AS #	32	1.8E-06	5 I		29
92	235	URANIUM	7.10E+08	A,G	5.3E-04	AS +	35	1.3E-08	SI		29
92	236	URANIUM	2.39E+07	A.G.E-	7.8E-04	AS *	32	4.6E=07	SI		29
92	238	URANIUM	4.50E+09	A,G,E-	8.2E-04	AS	32	2.4E-09	31 '		29
94	236	PLUTONIUM	2.85E+00	A,G,E-	3.7E-04	AS	8	8.0E-05	81	T.I	7
94	530	PLUTONIUM	0.645+01	A,G,E-	3.7E-04	AS	•	2.65-06	31	I,T	7
94	239	PLUTONIUM	2.44E+04	A.G.E-	3.7E-04	AS *	8	8.0E+09	31		7
94	240	PLUTONIUM	6.585+03	A,G,E-	3.7E-04	A5 +	8	3.2E-08	31		7
94	241	PLUTONIUM	1.35E+01	B-,A,G				1.5E-05	31		7
94	242	PLUTONIUM	3.80E+05	A,G	3.7E-04	AS	8	5.3E-10	SI		7
94	244	PLUTONIUM	7.60E+07	A	3.7E-04	AS	8	2.7E-12	SI		7

EXPLANATION OF ABBREVIATIONS USED IN TABLE 1.

- MAJOR RADIATIONS:

 A ALPHA PARTICLE
 B- BETA PARTICLE
 B+ POSITRON

 - E- CONVERSION ELECTRON
 G GAMMA RAY

COUNTING METHODS

- AS ALPHA SPECTROMETRY
 GS GAMMA SPECTROMETRY
 PC PROPORTIONAL COUNTER
 EE ELECTPOLYTIC ENRICHMENT
 NAA NEUTRON ACTIVATION ANALYSIS

- MASS SPECTROMETER METHOD:
 SI SURFACE IDNIZATION
 NSI NEGATIVE SURFACE IDNIZATION
 SG STATIL GAS
 HE HIGH ENERGY (ACCELERATOR)

MASS SPECTROMETER LIMITATIONS: (LISTED IN ORDER OF IMPORTANCE IN TABLE) C CHEMICAL INTERFERENCES I ISOBAR INTERFERENCES S STABLE ISOTOPE TAILING T HALF LIFE CONSIDERATIONS S RELATIVE COST IE IONIZATION EFFICIENCY

NOTES

- * ALPHA SPECTROMETRY DOES NOT RESOLVE THE ENERGY OF THESE ISOTOPES.

 ** M. S. SENS. GIVEN PER CC OF AIR BASED ON 1.14 PPM KRYPTON (ATMOSPHERIC ABUNDANCE).

 ** M. S. SENS. GIVEN PER GRAM OF SAMPLE BASED ON 7 PPM CESIUM (CRUSTAL ABUNDANCE).

- (CRUSTAL ABUNDANCE).

 M. S. SENS. GIVEN PER GRAM OF SAMPLE BASED ON 16PPM LEAD (CRUSTAL ABUNDANCE).

 M. S. SENS. GIVEN PER GRAM OF SAMPLE BASED ON 0.3PPM IODINE (CRUSTAL ABUNDANCE).

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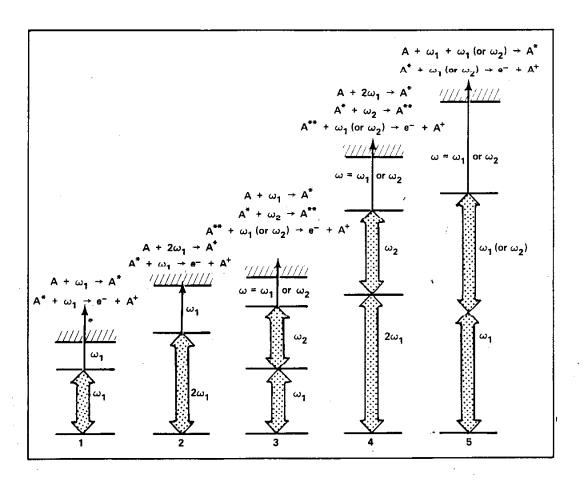


FIGURE 1. With These Five Laser Schemes all the Known Elements Except Helium and Neon can be Ionized. The Schemes are Classified here by a Notation Similar to that Used in Nuclear Physics.

(Taken from G. S. Hurst, et al., Reference 18)